

# PATENT ABSTRACTS OF JAPAN

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## (54) MOISTURE-CURABLE COMPOSITION

### (57)Abstract:

PROBLEM TO BE SOLVED: To provide a modified-silicone-based moisture-curable composition of which the stability of a cure catalyst is excellent.

SOLUTION: This composition mainly comprises (A) 100 pts.wt. silyl-group- containing organic polymer having at least one silicon atom bonded to a hydrolyzable group at the molecular end or in the side chain and (B) 0.1-10 pts.wt. cure catalyst, provided the catalyst is a mixture comprising 80-50 wt.% dialkyltin compound and 20-50 wt.% silicate compound.

## \* NOTICES \*

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1. This document has been translated by computer. So the translation may not reflect the original precisely.

2. \*\*\* shows the word which can not be translated.

3. In the drawings, any words are not translated.

## CLAIMS

[Claim 1a]

[Claim 1a] molecular terminal — or — a side chain — hydrolytic — a basis — having joined together — a silicon atom — one — a molecule — inside — at least — one — a piece — having — a silyl group — content — organicity — a polymer (—A—) 100 — a weight section — a curing catalyst — (—B—) 0.1 — ten — a weight section — a base component — a curing catalyst — a curing catalyst — a curing catalyst — a curing catalyst — a curing catalyst (B) is a mixture of 80 to 50 % of the weight of diallyl tin compounds, and 20 to 50 % of the weight of silicate compounds.

[Claim 2A] diallyl tin compound is general formula (1)  $R'_2SnX_2$  (1).

(Among a formula,  $R'_1$  is straight chain shape or a branched-chain alkyl group with 1-12 carbon atoms, and  $X$  Straight chain shape or a branched-chain alkoxy/ group with 1-18 carbon atoms, a basic chosen from a group which consists of straight chain shape with an ORGANO silicate group and 1-8 carbon atoms or a branched-chain acetoaceto-oid-alkyl-ester group, and an acetylacetone group — it is — the moisture curing nature constituent according to claim 1 which is a compound expressed.

[Claim 3A] silicate compound is general formula (2)  $2_nSi(OR^3)_{4-n}$  (2).

(Among a formula,  $R^2$  and  $R^3$  are straight chain shape or branched-chain alkyl groups of the carbon numbers 1-4 independently, and, respectively)  $n$   $R^2$  and  $R^3$  of an individual (4-n) may be the same, or may differ from each other in an integer of 0-3 — it is — the moisture curing nature constituent according to claim 1 which is a compound expressed or its hydrolyzate.

[Translation done.]





among these.

2002) In the moisture curing natura constituent of this invention, the content of a curing catalyst (B) has been 0.1–10 preferred weight action to silyl group content organicity polymer (A) 100 weight section, and has said less than range of hardening performance is [the quantity of a curing catalyst (B)]

to improve adhesion to a substrate. Can use publicly known various amino group substitution alkoxy silanes compounds or the condensate of those, and specifically, gamma-aminopropyl trimethoxysilane, gamma-aminopropyl triethoxysilane, N-(trimethoxysilylpropyl) ethylenediamine, delta-aminobutyl trimethoxysilane, N-bis(trimethoxysilylpropyl)ethylenediamine, these partial hydroxyates,

(003) Additive agents usually added by the hardenability constituent, such as a bulking agent, solvent, plasticizer, a hardening accelerator, a concrete retarder, a dropping inhibitor or an antiaging agent, and a pigment may be further added to the mixture during nature consequent of this invention. As a bulking agent, specifically For example, calcium carbonate, kaolin, etc., fumed silica, de-aeration treatment nature silica, silicic acid amorphous, hydrous silicon oxide clay, Calcination oily glass beads, aluminum nitrate, ball millon, glass fiber, asbestos, a glass filament grinding quartz, hematite, magnetite, organic bentonite, a little malloin, glass filler, asbestos, a titanium dioxide, aluminum alkoxide, aluminum hydroxide, a zinc oxide, magnesium oxide, a titanium dioxide, etc. are mentioned. Specifically as colorant iron oxide, carbon black, copper phthalocyanine blue, "Phthalomylene Green," etc. are mentioned. As a plasticizer, specifically Dodecyl phthalate, dioxybisphenols A, bisphenols B, Phthalic ester, such as butylenediethyl phthalates, diacetyl adipate, Succinic acid dicyclohexylphthalate, Diethylester succinate acid disoedeyl, diisododecyldioester, Glycid ether, such as aliphatic-carboxylic-alkoxy ester, polybutadiene glycol dimethyl acrylate, tetrahydrofuran ester, Epoxy system plasticizers, such as propoxypolyols, such as butyl cellosil and pentaerythrinit ester. Epoxy system plasticizers, such as triethyl phosphoric ester, such as tricresyl phosphate and triurenyl phosphates epoxidized soybean oil, and epoxyglycid stearic acid benzyl, a chlorinated paraffin, etc. are mentioned. Colloidal silica, etc. are mentioned. Organic benzoate, a chlorinated paraffin, etc. are mentioned. Colloidal silica, etc. are mentioned. Hydrogenation castor oil, a silicic acid amorphous, organic Bentonitic, colloidal silica, etc. are mentioned. As other additive agents, adhesion grant agents, such as phenol resin and an epoxy resin, an ultraviolet ray absorbent, radical chain inhibitors, a peroxide decomposition agent, various kinds of antiaging agents, etc. are mentioned.

[0030]

[0030] Although this invention is concretely explained based on an example below, the range of the invention is not limited by this.

2003). Electrolysis was performed in a three-necked round-bottomed flask equipped with a thermometer, a reflux condenser, and an agitator in the 500-mL 4-necked flask fitted with a nitrogen inlet. A 100-mL solution of 4-iodobenzonitrile (1.0 mol) in 200 mL of toluene and 40 mL of diethyl ether was added to the flask which was then stirred under a nitrogen atmosphere. A 40.8 g (0.2 mol) of diethylzinc solution, 52.1 g (0.2 mol) of *n*-butyllithium solution, and 200 g of toluene and it was made to react at 112 °C for 2 days, and after carrying out stirring drying of the generated water, toluene was distilled off under reduced pressure. The residue was then dissolved in 100 mL of toluene and 10 mL of diethyl ether. After decompression and light yellow transparent liquid A 92.4% (94% of yield) was obtained. This compound was checked generation of the diethyl tin (screw 2.2) with the disappearance of the absorption (3300–3400 cm<sup>-1</sup>) of O–H of alcohol and the advent of absorption (905 cm<sup>-1</sup>) of Sn–O–C bond.

(0032)In the same 4 mouth flask as the example 1 of example of manufacture 2 manufacture, 49.8 g (0.2 mol) of dibutyltin oxide, The acetic acid 24.0g (0.4 mol) and 200 g of toluene are taught. After carrying out reaction processing like the example 1 of manufacture, subsequently prepared 83.3 g (0.4 mol) of ethyl ortho silicate (tetraethoxysilane). It was made to react at 120 °\* for 3 hours.

Decompression distilling off of the generated ethyl acetate was carried out, and light yellow transparent liquid B 114.7g (97% of yield) was obtained. This compound has checked generation of the dilute/tin screw (TOMETOKISHI siliante) in FT-IR from the result of disappearance of absorption

[0033]

Measured value 40.8 ± 2.1 7.9 ± 20.1 Theoretical value 40.8 ± 2.1 8.9 ± 20.1 [003]/49.8 (g (0.2 mol) of diethyltin oxide, 40.0 g (0.4 mol) of acet/acetonates, and 200 g of toluene were taught to the same 4 smooth flask as the example 1 of example of manufacture, it reacted and processed (smooth flask as the example 1 of example of manufacture) was

http://www4iodf.inn.it/sgip/cgi-bin/transaction?atw=3A%2F%2Fwww4.ipd... 2010/03/03

obtained. This compound has checked generation of the dibutyl tin screw (acetylacetonato) with the solvent of absorption ( $547\text{ cm}^{-1}$ ) of  $\text{Sn-O-C}$  in FT-IR.

0.0035/49.8 (0.02 mol) of butyltin oxide, 52.1 g (0.4 mol) of ethyl acetate, and 200 g of toluene were taught to the same 4 mouth flask as the example 1 of manufacture 4 manufacture, it was reacted and processed like the example 1 of manufacture, and light yellow transparent liquid D 98.9g (9.7% of yield) was obtained. This compound has checked generation of the dibutyltin screw (ethylacetate) with the advent of absorption ( $550\text{ cm}^{-1}$ ) of  $\text{Sn-O-C}$  in FT-IR.

[0036] 49.8 g (0.2 mol) of dibutyltin oxide, 156.2 g (0.4 mol) of dioctyl phthalate, and 200 g of toluene were taught to the same 4 mouth flask as the example 1 of manufacture of example 5 manufacture, it were taught to the same 4 mouth flask as the example 1 of manufacture, and light yellow transparent liquid E 198.8 g (91% of yield) was obtained. This compound has checked generation of dibutyl tin dioctyl oxide with the solvent of absorption ( $550\text{ cm}^{-1}$ ) of  $\text{Sn}-\text{O}-\text{C}$  in FT-IR.

[0037] Instead of the dibutyltin oxide of the example 2 of example of manufacture 6 manufacture, 72.2 g (0.2 mmol) of dioctyl tin oxide was used, it reacted and processed by the same combination as the example 2 of manufacture, and light yellow fluid (136.5g (9% of yield) was obtained. This compound has checked generation of the dioctyl tin screw (T0RETIKOSILITE silicate) in FT-IR from the result of disappearance of absorption ( $1638\text{ cm}^{-1}$ ,  $1559\text{ cm}^{-1}$ ) of tin carbonyl, and the next ultimate analysis.

	C (%)	H (%)	O (%)	Si (%)	S (%)
Sample 1	55.2	4.8	38.5	1.2	0.3
Sample 2	56.1	4.9	37.8	1.1	0.2
Sample 3	57.0	5.0	37.0	1.0	0.1
Sample 4	58.5	5.2	35.5	0.8	0.0
Sample 5	59.8	5.4	34.2	0.6	0.0
Sample 6	61.2	5.6	32.8	0.4	0.0
Sample 7	62.5	5.8	31.5	0.2	0.0
Sample 8	63.8	6.0	30.2	0.1	0.0
Sample 9	65.0	6.2	28.8	0.0	0.0
Sample 10	66.2	6.4	27.5	0.0	0.0
Sample 11	67.5	6.6	26.2	0.0	0.0
Sample 12	68.8	6.8	24.8	0.0	0.0
Sample 13	70.0	7.0	23.5	0.0	0.0
Sample 14	71.2	7.2	22.2	0.0	0.0
Sample 15	72.5	7.4	20.8	0.0	0.0
Sample 16	73.8	7.6	19.5	0.0	0.0
Sample 17	75.0	7.8	18.2	0.0	0.0
Sample 18	76.2	8.0	17.0	0.0	0.0
Sample 19	77.5	8.2	15.8	0.0	0.0
Sample 20	78.8	8.4	14.5	0.0	0.0
Sample 21	80.0	8.6	13.2	0.0	0.0
Sample 22	81.2	8.8	12.0	0.0	0.0
Sample 23	82.5	9.0	10.8	0.0	0.0
Sample 24	83.8	9.2	9.5	0.0	0.0
Sample 25	85.0	9.4	8.2	0.0	0.0
Sample 26	86.2	9.6	7.0	0.0	0.0
Sample 27	87.5	9.8	5.8	0.0	0.0
Sample 28	88.8	10.0	4.5	0.0	0.0
Sample 29	90.0	10.2	3.2	0.0	0.0
Sample 30	91.2	10.4	2.0	0.0	0.0
Sample 31	92.5	10.6	0.8	0.0	0.0
Sample 32	93.8	10.8	0.0	0.0	0.0
Sample 33	95.0	11.0	0.0	0.0	0.0
Sample 34	96.2	11.2	0.0	0.0	0.0
Sample 35	97.5	11.4	0.0	0.0	0.0
Sample 36	98.8	11.6	0.0	0.0	0.0
Sample 37	100.0	11.8	0.0	0.0	0.0

Measured value 47.9, 5.3, 17.9, 8.1, 17.0. Theoretical—value 47.8, 8.9, 18.1, 8.0. 16.9, 0.0039. Instead of the diethyl tin oxide of the example 4 of example of manufacture 1 manufacture, 7.22 g (0.02 mol) of diethyl tin oxide was used. It reacted and processed by the same combination as the example 4 of manufacture, and light yellow fluid G 104.3g (96% of yield) was obtained. This compound has checked generation of the diethyl tin screw (ethylacetacetate) with the advent of absorption (547  $\text{cm}^{-1}$ ) of

[0400] Instead of the dibutyltin oxide of the example 2 of example of manufacture 8 manufacture, 33.0 g (0.2 mol) of dimethyl tin oxide was used, it reacted and processed by the same combination as the example 2 of manufacture, and the light yellow fluid H99.4g (98% of yield) was obtained. This compound has checked generation of the dimethyl tin screw (TORIETOKISHI silicate) in FT-IR from the result of disappearance of absorption (1638  $\text{cm}^{-1}$ , 1559  $\text{cm}^{-1}$ ) of in carbonyl, and the next

**[0041]** Measured value 32.7, 25.1, 11.1, 23.3 Theoretical-value 33.2, 7.1, 25.2(1.1) 23.4[0042]the tin compounds A-H obtained in examples 1-12 of an experiment; and the example 1 of comparative experiments - the examples 1-8 of the stability test of catalyst $\gamma$  manufacture - to 50 weight sections, respectively. What Examples 1, 5, 7, 9-12 of an experiment) mixed ethyl ortho silicate (tetraethoxysilane - 50 weight section, and tin compound A-D obtained in the examples 1-4 of manufacture - to 50 weight sections, respectively. What (examples 2, 4, 6, and 8 of an experiment) mixed ethyl silicate 40 (poly silicic acid acid ester by Tama Chemicals Co., Ltd. (4 - pentamer)) 50 weight section, and what does not add a tetraethoxysilane and the ethyl silicate 40 to tin compound A-H obtained in the examples 1-4 of manufacture as a comparative examination. What (comparative experiments) the tin compounds A-H obtained in the examples 1-4 of manufacture as the comparative experiments) are prepared in the same manner as described in the experimental part.

The state of the reaction mixture was observed at the time. The judgment of the state was performed based on the following standard. A result is shown in Table 1.

Q = clear state,

Measured value 33.2, 13.3, 25.1, 11.1, 23.3. Theoretical-value 33.2, 7.1, 25.21, 11.2, 23.3 (0.042) the tin compound A-H shown in examples 1–12 of an experiment and the example 1 of comparative experiments – the examples 1–8 of the stability test of catalytic manufacture – to 50 weight sections, respectively. What (examples 1, 3, 5, 7, 9–12 of an experiment) mixed ethyl ortho silicate (tetraethoxysilane) 50 weight section, and tin compound A-D obtained in the examples 1–4 of a manufacture – to 50 weight sections, respectively. What (examples 2, 4, 6, and 8 of an experiment) mixed ethyl silicate 40 glycol silicic acid ester by Tama Chemicals Co., Ltd. (4:0 + pentamer) 50 weight section. What and does not add a tetraethoxysilane and the ethyl silicate 40 tin compound A-H obtained in the examples 1–5 of a manufacture as a comparative example (examples 1–8 of comparative experiments) was put into the transparent glass bottle by the time. The judgment of the state temperature was made by the thermometer and the thermometer was placed in the bottle. The judgment of the state temperature was based on the following standard. A result is shown in Table 1.

Q. 2. The clear state.

\*\* Thickening or

x: Gelling or precipitate arose.

[0043]

[Table 1]

実施例	配合割合 (重量部)	試験				
		1日経	3日経	5日経	7日経	14日経
実施例1	硬化剤A 50	○	○	△	△	△
実施例2	硬化剤A 50	○	○	△	△	△
実施例3	硬化剤A 50	○	○	△	△	△
実施例4	硬化剤A 50	○	○	△	△	△
実施例5	硬化剤A 50	○	○	△	△	△
実施例6	硬化剤A 50	○	○	△	△	△
実施例7	硬化剤A 50	○	○	△	△	△
実施例8	硬化剤A 50	○	○	△	△	△
実施例9	硬化剤A 50	○	○	△	△	△
実施例10	硬化剤A 50	○	○	△	△	△
実施例11	硬化剤A 50	○	○	△	△	△
実施例12	硬化剤A 50	○	○	△	△	△
比較実施例1	硬化剤A 100	○	○	○	○	○
比較実施例2	硬化剤A 100	○	○	○	○	○
比較実施例3	硬化剤A 100	○	○	○	○	○
比較実施例4	硬化剤A 100	○	○	○	○	○
比較実施例5	硬化剤A 100	○	○	○	○	○
比較実施例6	硬化剤A 100	○	○	○	○	○
比較実施例7	硬化剤A 100	○	○	○	○	○
比較実施例8	硬化剤A 100	○	○	○	○	○
比較実施例9	硬化剤A 100	○	○	○	○	○
比較実施例10	硬化剤A 100	○	○	○	○	○
比較実施例11	硬化剤A 100	○	○	○	○	○
比較実施例12	硬化剤A 100	○	○	○	○	○
比較実施例13	硬化剤A 100	○	○	○	○	○
比較実施例14	硬化剤A 100	○	○	○	○	○
比較実施例15	硬化剤A 100	○	○	○	○	○
比較実施例16	硬化剤A 100	○	○	○	○	○
比較実施例17	硬化剤A 100	○	○	○	○	○
比較実施例18	硬化剤A 100	○	○	○	○	○
比較実施例19	硬化剤A 100	○	○	○	○	○
比較実施例20	硬化剤A 100	○	○	○	○	○
比較実施例21	硬化剤A 100	○	○	○	○	○
比較実施例22	硬化剤A 100	○	○	○	○	○
比較実施例23	硬化剤A 100	○	○	○	○	○
比較実施例24	硬化剤A 100	○	○	○	○	○
比較実施例25	硬化剤A 100	○	○	○	○	○
比較実施例26	硬化剤A 100	○	○	○	○	○
比較実施例27	硬化剤A 100	○	○	○	○	○
比較実施例28	硬化剤A 100	○	○	○	○	○
比較実施例29	硬化剤A 100	○	○	○	○	○
比較実施例30	硬化剤A 100	○	○	○	○	○
比較実施例31	硬化剤A 100	○	○	○	○	○
比較実施例32	硬化剤A 100	○	○	○	○	○
比較実施例33	硬化剤A 100	○	○	○	○	○
比較実施例34	硬化剤A 100	○	○	○	○	○
比較実施例35	硬化剤A 100	○	○	○	○	○
比較実施例36	硬化剤A 100	○	○	○	○	○
比較実施例37	硬化剤A 100	○	○	○	○	○
比較実施例38	硬化剤A 100	○	○	○	○	○
比較実施例39	硬化剤A 100	○	○	○	○	○
比較実施例40	硬化剤A 100	○	○	○	○	○
比較実施例41	硬化剤A 100	○	○	○	○	○
比較実施例42	硬化剤A 100	○	○	○	○	○
比較実施例43	硬化剤A 100	○	○	○	○	○
比較実施例44	硬化剤A 100	○	○	○	○	○
比較実施例45	硬化剤A 100	○	○	○	○	○
比較実施例46	硬化剤A 100	○	○	○	○	○
比較実施例47	硬化剤A 100	○	○	○	○	○
比較実施例48	硬化剤A 100	○	○	○	○	○
比較実施例49	硬化剤A 100	○	○	○	○	○
比較実施例50	硬化剤A 100	○	○	○	○	○
比較実施例51	硬化剤A 100	○	○	○	○	○
比較実施例52	硬化剤A 100	○	○	○	○	○
比較実施例53	硬化剤A 100	○	○	○	○	○
比較実施例54	硬化剤A 100	○	○	○	○	○
比較実施例55	硬化剤A 100	○	○	○	○	○
比較実施例56	硬化剤A 100	○	○	○	○	○
比較実施例57	硬化剤A 100	○	○	○	○	○
比較実施例58	硬化剤A 100	○	○	○	○	○
比較実施例59	硬化剤A 100	○	○	○	○	○
比較実施例60	硬化剤A 100	○	○	○	○	○
比較実施例61	硬化剤A 100	○	○	○	○	○
比較実施例62	硬化剤A 100	○	○	○	○	○
比較実施例63	硬化剤A 100	○	○	○	○	○
比較実施例64	硬化剤A 100	○	○	○	○	○
比較実施例65	硬化剤A 100	○	○	○	○	○
比較実施例66	硬化剤A 100	○	○	○	○	○
比較実施例67	硬化剤A 100	○	○	○	○	○
比較実施例68	硬化剤A 100	○	○	○	○	○
比較実施例69	硬化剤A 100	○	○	○	○	○
比較実施例70	硬化剤A 100	○	○	○	○	○
比較実施例71	硬化剤A 100	○	○	○	○	○
比較実施例72	硬化剤A 100	○	○	○	○	○
比較実施例73	硬化剤A 100	○	○	○	○	○
比較実施例74	硬化剤A 100	○	○	○	○	○
比較実施例75	硬化剤A 100	○	○	○	○	○
比較実施例76	硬化剤A 100	○	○	○	○	○
比較実施例77	硬化剤A 100	○	○	○	○	○
比較実施例78	硬化剤A 100	○	○	○	○	○
比較実施例79	硬化剤A 100	○	○	○	○	○
比較実施例80	硬化剤A 100	○	○	○	○	○
比較実施例81	硬化剤A 100	○	○	○	○	○
比較実施例82	硬化剤A 100	○	○	○	○	○
比較実施例83	硬化剤A 100	○	○	○	○	○
比較実施例84	硬化剤A 100	○	○	○	○	○
比較実施例85	硬化剤A 100	○	○	○	○	○
比較実施例86	硬化剤A 100	○	○	○	○	○
比較実施例87	硬化剤A 100	○	○	○	○	○
比較実施例88	硬化剤A 100	○	○	○	○	○
比較実施例89	硬化剤A 100	○	○	○	○	○
比較実施例90	硬化剤A 100	○	○	○	○	○
比較実施例91	硬化剤A 100	○	○	○	○	○
比較実施例92	硬化剤A 100	○	○	○	○	○
比較実施例93	硬化剤A 100	○	○	○	○	○
比較実施例94	硬化剤A 100	○	○	○	○	○
比較実施例95	硬化剤A 100	○	○	○	○	○
比較実施例96	硬化剤A 100	○	○	○	○	○
比較実施例97	硬化剤A 100	○	○	○	○	○
比較実施例98	硬化剤A 100	○	○	○	○	○
比較実施例99	硬化剤A 100	○	○	○	○	○
比較実施例100	硬化剤A 100	○	○	○	○	○

[0044] Examples 1-12, one to comparative examples 8 Examples 1, 2, 7, 8, and 11, and the comparative examples 1, 4, and 7 the catalyst after an end for stability test 21 day with the compounding ratio with the hardening catalyst. Comparative examples 2, 5, 6, and 8 show the catalyst after the end of stability test three months in various materials and Table 2. The room temperature of 25 °C. It was measured after loading in the thermostatic chamber of 60% of humidity, and a snap test (time until it becomes a letter of half-pulling), and tack free time (time until surface tackiness is lost) were measured. A result is shown in Table 2. The material in Table 2 expresses the following thing.

[0045] MS polymer S303: The polymer containing a hydrolytic silicon group (made by Kaneka Corp.)

NOKUPAKU NS-8: Antislip agent product made from Ouchi Shinko Chemical Industry

DOP : — diethyl phthalate A-1100 : Amino group substitution alkoxy silane compound (made by

Nippon Unicar)

[0046]

[Table 2]

[0047] The hardenability constituent (examples 1-12) which uses the catalyst which consists of the diallyl tin compound and allcinate compound of this invention so that clearly from Table 1 and Table 2. Compared with the hardenability constituent (comparative examples 1-8) which contains only a diallyl tin compound as a catalyst, it is stable in the air and it turns out that catalytic activity is not deactivated for a long period of time.

[0048]

[Effect of the invention] In the air, catalytic activity is not lost for a long period of time, but the moisture curing nature constituent of this invention is useful as a sealing agent, a coating agent, and elastic adhesives.

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[Translation done.]